

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Division of Patent Application Serial No. 09/303,405 of

IMAI et al

Atty. Ref.: 1417-348

Serial No. to be assigned

Group:

Filed: April 25, 2001

Examiner:

For: IRON COMPOUND CATALYST FOR INHIBITING GENERATION OF DIOXIN
AND INCINERATION PROCESS OF MUNICIPAL SOLID WASTE USING
THE SAME

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April 25, 2001

Assistant Commissioner for Patents
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

In order to place the above-identified application in better condition for
examination, please amend the application as follows:

IN THE SPECIFICATION:

Delete the current Abstract and replace it with the attached Abstract

IN THE CLAIMS:

Cancel claim 1.

Add the following new claims:

16. (New) An iron compound catalyst for inhibiting the generation of dioxin, comprising iron oxide particles, iron oxide hydroxide particles or mixture thereof and having a catalytic activity capable of converting at least 15 % of carbon monoxide into carbon dioxide when 2.8×10^{-4} mol of iron oxide particles obtained by heat-treating said iron compound catalyst in air at a temperature of 800°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor,

said iron oxide particles or said iron oxide hydroxide particles having an average particle size of 0.01 to 2.0 μm, a BET specific surface area of 0.2 to 200 m²/g, a phosphorus content of less than or equal to 0.02 % by weight, a sulfur content of less than or equal to 0.1 % by weight and a sodium content of less than or equal to 0.2 % by weight.

17. (New) An iron compound catalyst for inhibiting generation of dioxin according to claim 16, wherein the average particle size is 0.02 to 1.0 μm; the BET specific surface area is 0.5 to 100 m²/g and the phosphorus content is less than or equal to 0.005 % by weight.

18. (New) An iron compound catalyst for inhibiting generation of dioxin according to claim 16, wherein the catalytic activity capable of converting of carbon monoxide into carbon dioxide is at least 20 % when 2.8×10^{-4} mol of iron oxide particles obtained by heat-treating the iron compound catalyst in air at a temperature of 800°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor.

19. (New) An iron compound catalyst for inhibiting generation of dioxin according to claim 16, wherein said iron compound catalyst comprises aggregates comprising said iron oxide particles, said iron oxide hydroxide particles or mixture thereof,

said aggregates having a specific surface area of not less than 1.0 m²/cm³ when measured under a feed pressure of 1 bar in a dry granulometer, and an average particle size (D50) of 50 % of a total volume thereof, of up to 8.0 μm.

20. (New) An iron compound catalyst for inhibiting generation of dioxin according to claim 19, wherein the specific surface area of said aggregates is at least 1.2 m²/cm³ when measured under a feed pressure of 1 bar in a dry granulometer, and the average particle size (D50) is up to 7.0 μm.

21. (New) An iron compound catalyst for inhibiting generation of dioxin according to claim 19, wherein said aggregates comprise said iron oxide particles, said iron oxide hydroxide particles or mixture of said iron oxide particles and said iron oxide hydroxide particles having an average particle size of 0.02 to 1.0 μm , a BET specific surface area of 0.5 to 100 m^2/g , a phosphorus content of less than or equal to 0.005 % by weight, a sulfur content of less than or equal to 0.1 % by weight and a sodium content of less than or equal to 0.2 % by weight.

22. (New) An incineration process of a municipal solid waste, comprising:
spray-introducing said iron compound catalyst as defined in claim 16 in an amount of 0.01 to 5.0 % by weight per hour based on the weight of a dry municipal solid waste, into a combustion chamber of an intermittently operated incinerator by a gas carrying method to contact said iron compound catalyst with a combustion gas.

23. (New) An incineration process according to claim 22, wherein said gas is an air or a nitrogen, and the amount of said gas is 1 to 20 % by volume based on the total volume of combustion gas fed into said combustion chamber.

24. (New) An incineration process according to claim 22, wherein said iron compound catalyst comprises the aggregates of said iron oxide particles, said iron oxide hydroxide particles or mixture thereof,

said aggregates having a specific surface area of not less than $1.0 \text{ m}^2/\text{cm}^3$ when measured under a feed pressure of 1 bar in a dry granulometer, and an average particle size (D50) of 50 % of a total volume thereof, of up to $8.0 \text{ }\mu\text{m}$.

25. (New) An incineration process according to claim 22, wherein the amount of said iron compound catalyst spray-added is 0.1 to 1.0 % by weight per hour based on the weight of the dry municipal solid waste.

26. (New) An incineration process of municipal solid waste, comprising:
spray-adding said iron compound catalyst as defined in claim 16 into the combustion chamber of the intermittently operated incinerator by a gas carrying method while supplying a secondary gas toward a tip end of combustion flame in the combustion chamber of the intermittently operated incinerator to uniformly disperse said iron compound catalyst in the combustion chamber, thereby contacting said iron compound catalyst with a combustion gas.

27. (New) An incineration process according to claim 26, wherein said secondary gas is air, a nitrogen gas or a combustion exhaust gas.

28. (New) An incineration process according to claim 26, wherein the amount of said secondary gas is 1 to 40 % by volume based on the total volume of the combustion air fed into said combustion chamber.

29. (New) An iron compound catalyst for inhibiting generation of dioxin, comprising aggregates comprising iron oxide particles, iron oxide hydroxide particles or mixture thereof and having a specific surface area of not less than $1.0 \text{ m}^2/\text{cm}^3$ when measured under a feed pressure of 1 bar in a dry granulometer, and an average particle size (D50) of 50 % of a total volume thereof, of up to $8.0 \text{ }\mu\text{m}$, and

having a catalytic activity capable of converting at least 15 % of carbon monoxide into carbon dioxide when 2.8×10^{-4} mol of iron oxide particles obtained by heat-treating said iron compound catalyst in air at a temperature of 800°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of $42,400 \text{ h}^{-1}$ in an inert gas atmosphere using a pulse catalytic reactor.

said iron oxide particles or said iron oxide hydroxide particles having an average particle size of 0.01 to $2.0 \text{ }\mu\text{m}$, a BET specific surface area of 0.2 to $200 \text{ m}^2/\text{g}$, a phosphorus content of less than or equal to 0.02 \% by weight, a sulfur content of less than or equal to 0.1 \% by weight and a sodium content of less than or equal to 0.2 \% by weight.

30. (New) An incineration process of a municipal solid waste, comprising:

spray-introducing said iron compound catalyst as defined in claim 16 or 19 in an amount of 0.01 to 5.0 \% by weight per hour based on the weight of a dry municipal solid

waste, into a combustion chamber of an intermittently operated incinerator by a gas carrying method wherein a gas is an air or a nitrogen, and the amount of said gas is 1 to 20 % by volume based on the total volume of combustion gas fed into said combustion chamber, to contact said iron compound catalyst with a combustion gas.

31. (New) An incineration process of municipal solid waste, comprising:

spray-adding said iron compound catalyst as defined in claim 16 or 19 into the combustion chamber of the intermittently operated incinerator by a gas carrying method wherein a gas is an air or a nitrogen, and the amount of said gas is 1 to 20 % by volume based on the total volume of combustion gas fed into said combustion chamber, while supplying a secondary gas of air, a nitrogen gas or a combustion exhaust gas toward a tip end of combustion flame in the combustion chamber of the intermittently operated incinerator to uniformly disperse said iron compound catalyst in the combustion chamber, thereby contacting said iron compound catalyst with a combustion gas,

the amount of said secondary gas being 1 to 40 % by volume based on the total volume of the combustion air fed into said combustion chamber.

Delete the Abstract and insert the abstract attached as a separate page.

REMARKS

This application is a division of Serial No. 09/303,405 filed May 3, 1999 which contains allowed claims directed to an incineration process.

Please note the Information Disclosure Statement filed with this amendment.

The above claims are directed to an iron compound catalyst for inhibiting the generation of dioxin composed of iron oxide particles, iron oxide hydroxide particles or a mixture of iron oxide particles and iron oxide hydroxide particles. The catalyst is characterized by its activity as a catalyst and its physical and chemical properties:

(1) it has a catalytic activity capable of converting not less than 15 % of carbon monoxide into carbon dioxide when 2.8×10^{-4} mol of iron oxide particles obtained by heat-treating the iron compound catalyst in air at a temperature of 800°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor;

(2) the catalyst particles have

- (i) an average particle size of 0.01 to 2.0 μm,
- (ii) a BET specific surface area of 0.2 to 200 m²/g,
- (iii) a phosphorus content of not more than 0.02 % by weight,
- (iv) a sulfur content of not more than 0.1 % by weight, and
- (v) a sodium content of not more than 0.2 % by weight.

This invention also includes a process for incinerating municipal solid waste by spraying the iron compound catalyst as set forth above in an amount of 0.01 to 5.0 % by weight per hour based on the weight of dry municipal solid waste, into a combustion chamber of an intermittently operated incinerator using a gas carrying method to contact the iron compound catalyst with a combustion gas.

By using the iron compound catalyst of the invention to inhibit generation of dioxin, in the incineration of municipal solid waste in an intermittently operated incinerator, such as mechanical batch incinerators or semi-continuous incinerators, complete combustion of the municipal solid waste and decomposition dioxin precursors can be achieved. The process inhibits generation of dioxin due to the memory effect upon low-temperature combustion at start-up or shut-down of the incinerator.

For example, (a) upon start-up of the incinerator, the concentration of dioxin in the combustion exhaust gas can be reduced to not more than 10 ngTEQ/Nm³, and during steady operation of the incinerator, the concentration of dioxin in the combustion exhaust gas can be reduced to not more than 8.0 ngTEQ/Nm³; (b) upon start-up of the incinerator, the concentration of carbon monoxide in the combustion exhaust gas can be reduced to not more than 120 ppm and during steady operation of the incinerator, the concentration of carbon monoxide in the combustion exhaust gas can be reduced to not more than 50 ppm; and (c) upon start-up of the incinerator, the concentration of hydrogen chloride in the combustion exhaust gas can be reduced to not more than 60 ppm and during steady operation of the incinerator, the concentration of hydrogen chloride in the combustion exhaust gas can be reduced to not more than 40 ppm.

Applicants discuss below the relevant information contained in the documents cited during examination of the parent application and also explain how the new claims presented above distinguish from the disclosures of the documents cited either individually or in combination. In addition, applicants refer to information and data contained in two attachments to this response, Table 1 and Table 2 presented in the form of a declaration of Mr. T. Matsui made September 21, 2000.

US Patent No. 5,744,690 (Yokoyama et al) discloses a method of incinerating combustible wastes by incinerating the combustible wastes in the presence of a calcium compound and iron compound particles selected from iron oxide particles, ferric oxide hydroxide particles and mixed particles thereof. Yokoyama et al also disclose the use of

spindle-shaped goethite particles of BET specific surface area: 80 m²/g, average major axial diameter: 0.3 μm (in Example 1).

However, as seen from the attached data the content of sulfur is 0.15 % by weight and the content of sodium is 0.28 % by weight, which fall outside of the range of applicants' claims. So, the goethite particles used in Example 1 of Yokoyama et al are different from those of the present invention.

As a result, conversion of carbon monoxide into carbon dioxide in case of using goethite aggregates used in Example 1 of Yokoyama et al, is 1.5 %, which is inferior to that of the present invention, falls outside of the range achieved by the present invention.

US Patent No. 4,979,447 (Farrar) discloses a process for improving combustion efficiency in a coal-fired furnace while maintaining acceptable levels of NO_x emissions from such combustion. The coal-fired furnace has at least one site for introducing a pulverized coal into the furnace to develop and maintain a fireball in the furnace and at least one site for introducing air to support combustion of the pulverized coal in the fireball. The process includes introducing at least one site a useful amount of an additive composition for measurably improving combustion efficiency while maintaining acceptable levels of NO_x emissions by controlling excess air requirements to support burning of the pulverized coal. Thus Farrar relates to additive compositions for improving combustion efficiency in coal-fire furnaces and other related types of equipment.

In contrast, the primary reference Yokoyama et al relates to a method of incinerating combustible wastes as does the present invention. Yokoyama et al is in a different field of endeavor from that of Farrar, and therefore, can not be properly combined with Yokoyama et al.

Further, since the iron compound catalyst of the present invention is different from the goethite particles used in Yokoyama et al, even if Farrar is combined with Yokoyama et al, one of ordinary skill in the art would not reasonably expect inhibition of dioxin

generation due to a memory effect upon low-temperature combustion at start-up or shut-down of incinerators.

US Patent No. 4,495,874 (Greskovich et al) discloses a method for combusting high ash content coal in a furnace having a firebox and at least one injection nozzle having a bore for a primary airstream and an annular passage for a secondary airstream, the nozzle communicating with the firebox. The method includes (a) injecting a primary airstream containing pulverized coal which is at least 7 wt % ash and a secondary airstream through the injection nozzle into the flame zone of the furnace firebox and combusting the coal to yield a flame of combustion, and (b) injecting a sufficient amount of oxygen into the secondary airstream such that an oxygen enriched secondary airstream emanates from the injection nozzle and the oxygen concentration of the combined primary and secondary air streams is 22 to 25 vol %. The result is that the amount of ash fines less than one micron in diameter is reduced.

Like Farrar, Greskovich et al is concerned with a process for combusting pulverized coal which is fed into a furnace firebox with a primary airstream which is also supplied with a secondary airstream. By contrast, Yokoyama et al relates to a method of incinerating combustible wastes. Clearly Yokoyama et al invention is in a different field from that of Greskovich et al, and therefore, cannot be properly combined with Greskovich et al.

Further, since the iron compound catalyst of the present invention is different from the goethite particles used in Yokoyama et al, even if Farrar and/or Greskovich et al are combined with Yokoyama et al, one of ordinary skill in the art would still not be able to sufficiently and effectively inhibit dioxin generation due to a memory effect upon low-temperature combustion at start-up or shut-down of an incinerator.

An examination of the above claims on the merits is awaited.

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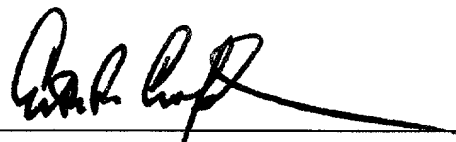
To assist the examiner attached is a copy of the Matsui Declaration filed September 28, 2000 in the parent application.

Attached hereto is a marked-up version of the changes made to the specification by the current amendment. The attached page is captioned "**Version With Markings To Show Changes Made.**"

Respectfully submitted,

NIXON & VANDERHYE P.C.

By: _____



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VERSION WITH MARKINGS TO SHOW CHANGES MADE
IN THE ABSTRACT:

ABSTRACT OF THE DISCLOSURE:

An iron compound catalyst for inhibiting the generation of dioxin of the present invention, comprise iron oxide particles, iron oxide hydroxide particles or mixed particles thereof having a catalytic activity capable of converting not less than 15 % of carbon monoxide into carbon dioxide when 2.3×10^{-4} mol of iron oxide particles obtained by heat-treating said iron compound catalyst in air at a temperature of 300°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of $42,400 \text{ h}^{-1}$ in an inert gas atmosphere using a pulse catalytic reactor, said iron oxide particles or said iron oxide hydroxide particles having an average particle size of 0.01 to 2.0 μm , a BET specific surface area of 0.2 to 200 m^2/g , a phosphorus content of not more than 0.02 % by weight, a sulfur content of not more than 0.6 % by weight and a sodium content of not more than 0.5 % by weight. Such an iron compound catalyst enables complete combustion of the municipal solid waste and decomposition of dioxin precursors even at a low combustion temperature in intermittently operated incinerators such as mechanical batch incinerators or semi-continuous incinerators, and can inhibit the generation of dioxin due to a memory effect upon low-temperature combustion at the start-up or shut-down of the incinerators, without large-scale incinerator renovation or plant and equipment investment.

ABSTRACT OF THE DISCLOSURE

An iron compound catalyst for inhibiting the generation of dioxin contains iron oxide particles, iron oxide hydroxide particles or mixed particles having a catalytic activity capable of converting not less than 15% of carbon monoxide into carbon dioxide when 2.8×10^{-4} mol of iron oxide particles obtained by heat-treating the iron compound catalyst in air at a temperature of 800°C for 15 minutes, are instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor, the iron oxide particles or the iron oxide hydroxide particles have an average particle size of 0.01 to 2.0 μm, a BET specific surface area of 0.2 to 200 m²/g, a phosphorus content of not more than 0.02% by weight, a sulfur content of not more than 0.6% by weight and a sodium content of not more than 0.5% by weight. This iron compound catalyst enables complete combustion of municipal solid waste and decomposition of dioxin precursors even at a low combustion temperature in intermittently operated incinerators such as mechanical batch incinerators or semi-continuous incinerators, and can inhibit dioxin generation of due to a memory effect upon low-temperature combustion at the start-up or shut-down of the incinerators, without large-scale incinerator renovation or plant and equipment investment.